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The goals of this program were investigate fundamental femtosecond-timescale phenomena in photonic materials and device structures, and to evaluate and demonstrate the potential of these phenomena for device applications. Significant progress was made in several areas, in the development of new solid-state femtosecond laser and fiber lasers, and in the application of femtosecond optics to studies of novel semiconducting and superconducting materials and waveguide structures.

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FINAL REPORT

Air Force Office of Scientific Research
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FEMTOSECOND PHOTONICS: FUNDAMENTAL PHENOMENA AND DEVICE BEHAVIOR

Principal Investigators

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Objectives

To investigate fundamental femtosecond-timescale phenomena in photonic materials and device structures. To evaluate the potential of these phenomena for device applications. To invent and demonstrate new devices for ultrafast optical applications.

Status and Accomplishments

Over the course of this contract we have made significant progress in several areas, both in the development of new femtosecond lasers and techniques and in the application of femtosecond optics to studies of novel materials and material structures, some of these key advances are summarized in sections entitled:

A. Ultrashort Pulse Generation in Solid State Lasers
B. Flashlamp-Pumped Ti:Al_2O_3 Laser Modelocking
C. Broad Spectrum Solid State Laser Development
D. Modelocking of Fiber Lasers
E. Efficient Frequency-Doubling of a Stretched-Pulse Fiber Laser
F. Wavelength Shifting in Passive InGaAsP/InP Quantum-well Waveguides
G. Femtosecond Optical Nonlinearities in ZnSe and Characterization of ZnSe/GaAs Heterostructures
H. Femtosecond Continuum Studies of PbTe Quantum Dots at 1.5 μm
I. Ultrafast Dynamics of K_3C_{60} and Rb_3C_{60} in the Normal and Superconducting Phase
J. Carrier Dynamics in InGaAs Strained Layer Diodes
K. Multi-wavelength Studies of The Nonlinear Index of Refraction in Narrow Bandgap Semiconductors

studies have indicated that these materials may be appropriate for the passive modelocking of flashlamp pumped Ti:Al_2O_3.

C. Broad Spectrum Solid State Laser Development

Numerous studies, including the investigation of ultrafast carrier relaxation processes in semiconductors and semiconductor devices and biomedical imaging, require the availability of broad spectral coverage in the infrared and near infrared wavelength range. Several broad bandwidth, tunable solid state sources have been developed, most notably Ti:Al_2O_3 (titanium sapphire), Cr:Mg_2SiO_4 (Cr:Forsterite), and Cr:YAG. The combination of these three sources alone can, in principle, cover the entire range from 0.65 μm to 1.65 μm. However, of these sources only titanium sapphire has been demonstrated to tune throughout its fluorescent emission range. This fact is largely due to presence of excited state absorption, parasitic absorption and poor figure of merit in the chromium doped crystals.

In this program we have explored two approaches to enhancing the wavelength coverage of the three laser crystals listed above. The first approach involves the development of novel pumping strategies which allow the use of shorter laser crystals and thereby reduce the deleterious effects of excited state and parasitic absorption. The second approach has explored new crystal growth procedures which may allow the development of more strongly doped and higher figure of merit crystals.

Fluorescent emission from Cr:Forsterite suggests the possibility to produce modelocked laser pulses in the wavelength range of 1.05 μm to 1.35 μm. However, an overlap with the absorption band on the short wavelength end of the spectrum and the presence of water absorption lines near 1.35 μm has limited the tuning range for modelocked operation. The convenience of pumping with the standard 1.06 μm Nd:YAG lasers, where the absorption of Cr:Forsterite is relatively weak, dictates the use of long laser crystals which preclude tuning below 1.18 μm. We have constructed a Nd:YAG pumped KLM Cr:Forsterite laser and have demonstrated tuning over the range 1.18 μm to 1.32 μm. This laser produces modelocked pulse durations of 30 fs and is currently being used for short coherence length biomedical imaging.

In collaboration with the International Laser Center, in Minsk, Belarus, we have investigated new crystal growth procedures which may allow the development of more strongly doped Cr:Forsterite crystals. Higher doping will allow the use of shorter laser crystals than previously used and should result in both greater tuning range and the production of shorter modelocked pulse durations. To evaluate this potential, we have investigated the spectroscopy and laser action of Cr:Forsterite crystals grown along the crystallographic c-axis, comparing their performance to the standard crystal growth which progresses along the a-axis. Our preliminary investigation focused on crystals from the two growth procedures having equivalent concentrations of chromium ions and have indicated that this new crystal growth process can produce laser gain media with performance comparable to that of the standard crystals.

D. Modelocking of fiber lasers

In our laboratory, we have pursued successfully the generation of modelocked pulse trains from additive pulse modelocked (APM) fiber lasers. The polarization in the fiber is made elliptical, and rotation of the polarization ellipse via the Kerr nonlinearity, followed by a polarizer, can produce intensity dependent loss. Effective saturable absorber action is produced resulting in the generation of modelocked trains of pulses, without the use of any other modulation mechanism.

short as 70 fsec, but due to the lower efficiency of gratings (~30%) frequency doubling was undertaken with prism compression.

By focusing the compressed fundamental pulses onto a 1 cm long AR-coated BaB_2O_4 (BBO) crystal, frequency-doubled powers as high as 8.7 mW were achieved, corresponding to 10% conversion efficiency and pulse energies of up to 270 pJ. The frequency-doubled pulse width was 86 fsec and the spectrum was 7.3 nm wide centered at 771 nm, resulting in a time-bandwidth product of 0.32, which is near transform-limited, assuming secant hyperbolic pulse shapes. These frequency-doubled pulse pulse energies are sufficient to seed high-repetition-rate regenerative amplifiers and avoid amplified spontaneous emission background.

Frequency doubling was also performed with a 1.5 mm $KNBO_3$ (potassium niobate) crystal and with a 7 mm LiB_3O_5 (LBO) crystal. Potassium niobate has a nonlinear coefficient which is two orders of magnitude larger than BBO, and we obtained conversion efficiency of 1.5% with this short crystal. We obtained 6% conversion efficiency with LBO, which is advantageous because it can be temperature tuned for 90-degree phase matching with no spatial walk-off. In both crystals, through the frequency-doubled pulse widths were > 170 fsec, indicating that although high efficiencies were obtained with short crystals, the smaller phase-matching bandwidth of these crystals would not allow efficient doubling of 100 fsec pulses.

Frequency-resolved optical gating (FROG), which allows the direct determination of the intensity and phase of an ultra-short pulse, was used to better characterize both the fundamental and frequency-doubled pulses and to explore why the doubled pulse spectrum was offset from the center of the fundamental spectrum. FROG data on the 1.55 μm pulse was taken by measuring the spectrum of the second harmonic generation (SHG) as the pulse delay was changed in a background-free autocorrelation configuration. The FROG trace indicated that the lower-frequency components are contained in the pulse wings; thus efficient frequency doubling occurs at shorter wavelengths. This explains why the SHG spectrum is centered at 771 rather than 776 nm, which would be predicted based on the first moment of the fundamental spectrum. We confirmed that our FROG data accurately represented the pulse by comparing the experimental 1.55 μm spectrum with the Fourier transform of the calculated pulse intensity and phase from the calculated FROG fit. There was excellent agreement between the two spectra, indicating that no filtering occurred in the pulse compression or nonlinear crystal.

The frequency-doubled pulses were also characterized with an SHG FROG measurement. The FROG trace was round and smooth, indicating that the pulse was near transform-limited with flat phase across the entire pulse. Amplitude noise measurements were performed on the frequency-doubled pulses and compared with those of a commercial Ti:sapphire laser pumped by an argon-ion laser. The amplitude fluctuations on the frequency-doubled pulses rolled off more quickly and were at -138 dBc/Hz at 100 kHz. The Ti:sapphire noise is typically at least 40 dBc/Hz higher over the range 3 - 100 kHz.

F. Wavelength Shifting in Passive InGaAsP/InP Quantum-well Waveguides

We have developed an analytical theory for nonlinear conversion by four-wave mixing (FWM) in the presence of two-photon absorption, and demonstrated its predictions experimentally. Theoretically, we have shown that while nonlinear loss enhances small-signal FWM, it places a fundamental upper limit on the conversion efficiency. Expressions for the optimum input pump intensity and associated maximum conversion were obtained, and optimum waveguide lengths determined. Experimentally, we used picosecond pulses from two synchronized color-center

conduction band offset at the interface. These results demonstrate that band bending due to interface charges must be taken into account for any study of the effect of growth and nucleation conditions on this interface and for understanding carrier transport in ZnSe based devices using GaAs substrates.

Additional pump-probe experiments were performed to extend the conventional photoreflectance measurements. The mechanism of photoreflectance techniques is screening of built-in surface fields by surface trapping of minority carriers. This reduction in the electric field changes the Franz-Keldysh contributions to the refractive index and, therefore, the sample reflectivity. Pump-probe experiments were performed to time resolve these subpicosecond trapping dynamics for the first time. Both the intensity and spectral dependence of the reflectivity signals support this interpretation. A simple model is proposed for explaining these results which shows that this technique may be used as a non-contact method for measuring surface trap densities and recombination velocities. Extension of this technique to the technologically important Si/SiO_2 interface would be straightforward due to the bandstructure transitions used.

H. Femtosecond Continuum Studies of PbTe Quantum Dots at 1.5 μm

With their strong quantum confinement semiconductor quantum dots in glass matrices offer the potential of application as efficient nonlinear optical elements. Previous work has shown that Cd-compound nanocrystallites exhibit strong optical nonlinearities in the visible. The lower bandgap of PbTe provides opportunity to produce quantum dots absorbing in the important telecommunication band at 1.5 μm.

We have used our recently demonstrated a source of femtosecond continuum around 1.5 μm to investigate optical nonlinearities of PbTe nanocrystallites in a glass matrix. The quantum dots (obtained through collaboration with C. L. Cesar at UNICAMP in Campinas, Brazil) are produced in a glass matrix by a heat treatment close to the softening temperature of the glass. The mean radii were controlled by annealing time and temperature, and the quantum confined energy level was varied from 1300 to 1800 nm, respectively.

For our pump wavelength of 1.52 μm, the different samples make possible pump-probe experiments over a range of above and below band excitations. Induced transmission is observed both above and below the excitation wavelength within the response time of our system, consistent with ultrafast screening and carrier-carrier scattering. Partial recovery, particularly of the lowest electronic states, occurs on a timescale of 10 ps. Similar partial recovery in CdS quantum dots has been explained previously as due to trapping by surface states. Complete electron-hole recombination follows on a longer timescale. Experiments to further elucidate these dynamics are underway.

I. Ultrafast Dynamics of K_3C_{60} and Rb_3C_{60} in the Normal and Superconducting Phase

The transient optical response of high-T_C and metallic superconductors to impulsive optical excitation by sub-picosecond light pulses gives important information on the dynamics of Cooper pair breaking and recombination. We have recently discussed photo-induced changes. For this reason we have performed pump-probe studies on superconducting potassium- and rubidium-doped C_{60} films at low temperature. As we have reported previously, the sensitivity of these

Gain depletion throughout the investigated spectral region was observed. For time delays longer than 1 ps, the gain partially recovers as the temperature reaches equilibrium with the lattice. The residual gain changes are produced by the decrease in the carrier population and recover on a much longer time scale.

In collaboration with theoretical physicists, G.D. Sanders and C.J. Stanton, at the University of Florida, Gainsville, detailed theoretical models for the gain dynamics for InGaAs strained layer diodes were developed. Calculations of the femtosecond gain dynamics in InGaAs/AlGaAs strained-layer single-quantum-well diode lasers were compared to experiments which used a multiple-wavelength pump probe technique. In the model developed, transient gain and differential transmission are computed in a multiband effective mass model including biaxial strain, valence subband mixing, and scattering both within and between subbands. The transient photogeneration of electron-hole pairs by the pump pulse and subsequent relaxation of carriers by both polar optical phonon scattering and carrier-carrier scattering are calculated within a Boltzmann equation framework. A relaxation approximation for the carrier-carrier scattering is made and the coupled Boltzmann equation are solved using an adaptive Runge-Kutta technique. This formalism aids the full understanding of the obtained experimental results and may aid future device design.

K. Multi-wavelength Studies of The Nonlinear Index of Refraction in Narrow Bandgap Semiconductors

The nonlinear optical properties of semiconductor materials are of great interest because of their role as components of various optical semiconductor devices. The dominant nonlinear contribution to the index of refraction is the third order nonlinear response of the material. It is important for various applications including optical limiting, all optical switching and modulation as well as for frequency modulation behavior of diode lasers.

Theory predicts the scaling of nonlinear index with semiconductor bandgap, E_g, as E_g^{-4}. For this reason, narrow bandgap semiconductor materials, such as InAs, GaSb, etc. are of particular importance. To date, most previous investigations of nonlinear index effects have used fixed wavelength or narrowly tunable laser sources and inferred the scaling behavior and wavelength dependence by performing experiments in different materials systems. Tunability constraints have been especially severe for these experiments which require high intensity short laser pulses. Using the Vanderbilt Free Electron Laser as a broadly tunable, high intensity source we have initiated the first systematic study of wavelength dependence of nonlinear index in narrow gap semiconductors.

There exist several techniques for measuring the nonlinear index. One of the simplest and most sensitive is Z-scan. It uses nonlinear propagation in order to measure the integrated nonlinear index. A high intensity laser beam is focused onto a material and the position of the material is scanned in the axial or Z direction. Scanning the Z position varies the focusing parameters of the beam in the nonlinear material and changes the propagation of the beam in the far field. Changes in the divergence of the beam are measured by measuring the transmission through an aperture positioned in the far field region behind the sample. From these transmission changes, the sign and magnitude of the nonlinear index can be calculated.

In collaboration with Prof. Norman Tolk's group at Vanderbilt University, we have used the Vanderbilt FEL to perform Z-scan experiments in InAs, GaSb and Ge. The Vanderbilt FEL is a high intensity laser source continuously tunable in the wavelength region from 2 to 10 microns

2. C.R. Doerr, W.S. Wong, H.A. Haus, and E.P. Ippen, "Additive-pulse Mode-locking/Limiting Storage Ring," Opt. Lett. **19**, 1747-1749 (1994).

3. C.R. Doerr, H.A. Haus, and E.P. Ippen, "Asynchronous soliton modelocking," Opt. Lett. **19**, 1958-1960 (1994).

4. K. Tamura, L.E. Nelson, H.A. Haus, and E.P. Ippen, "Soliton versus non-soliton operation of fiber ring lasers," Appl. Phys. Lett. **62**, 149-151 (1994).

5. C.-K. Sun, F. Vallee, L. H. Acioli, E. P. Ippen, and J. G. Fujimoto, "Femtosecond-tunable measurement of electron thermalization in gold," Phys. Rev. B **50**, 337-348, November 1994.

6. B. Bouma, A. Gouveia-Neto, J.A. Izatt, J. Russell, R. Sierra, U. Keller, and J.G. Fujimoto, "Hybrid modelocking of a flashlamp pumped Ti:Al_2O_3 Laser," Opt. Lett. **19**, 1858-1860 (1994).

7. M. Ramaswamy-Paye and J.G. Fujimoto, "Compact dispersion-compensating geometry for Kerr-lens modelocked femtosecond lasers," Opt. Lett. **19**, 1756-1758 (1994).

8. H.A. Haus, E.P. Ippen, and K. Tamura, "Additive pulse modelocking in fiber lasers," IEEE J. Quant. Electron. **30**, 200-208 (1994).

9. K. Tamura, C.R. Doerr, L. Nelson, H.A. Haus, and E.P. Ippen, "Technique for obtaining high-energy ultrashort pulses from an additive-pulse mode-locked erbium-doped fiber ring laser," Opt. Lett. **19**, 46-48 (1994).

10. E.P. Ippen, "Principles of passive mode locking," Appl. Phys. B **58**, 159-170 (1994).

11. F.I. Khatri, G. Lenz, J.D. Moores, H.A. Haus, and E.P. Ippen, "Extension of coupled-cavity additive pulse mode-locked laser theory," Opt. Commun. **110**, 131-136 (1994).

12. K.L. Hall, G. Lenz, A.M. Darwish, and E.P. Ippen, "Subpicosecond gain and index nonlinearities in InGaAsP diode lasers", Opt. Commun. **111**, 589-612 (1994).

13. K. Tamura, E.P. Ippen, and H.A. Haus "Optimization of filtering in soliton fiber lasers," IEEE Photonic Tech. Lett. **6**, 1433-1435 (1994).

14. H.A. Haus, K. Tamura, L.E. Nelson, and E.P. Ippen, "Stretched-pulse additive pulse mode-locking in fiber lasers: Theory and xxperiment," IEEE J. Quant. Electron. **31**, 591-598 (1995).

15. D.J. Dougherty, F.X. Kartner, H.A. Haus, and E.P. Ippen, "Measurement of the Raman gain spectrum of optical fibers," Opt. Lett. **20**, 31-33 (1995).

16. L.E. Nelson, E.P. Ippen, and H.A. Haus, "Broadly tunable sub-500 fs pulses from an additive-pulse mode-locked Thulium-doped fiber ring laser," Appl. Phys. Lett. **67**, 19-21 (1995).

17. C.-K. Sun, B. Golubovic, J.G. Fujimoto, H.K. Choi, and C.A. Wang, "Heterodyne nondegenerate pump-probe measurement technique for guided wave devices," Opt. Lett. **20**, 210-212 (1995).

Wong, and M. Haner, "All-optical network consortium ultrafast TDM networks", IEEE J. Selec. Areas in Commun. **54**, 999-1013 (1996).

33. A.M. Darwish, E.P. Ippen, H.Q. Le, J.P. Donnelly, and S.H. Groves, "Optimization of four-wave mixing conversion efficiency in the presence of nonlinear loss," Appl. Phys. Lett. **69**, 737-739 (1996).

34. B. Bouma and J.G. Fujimoto, "Compact Kerr-lens-mode locked resonators," Opt. Lett. **21**, 134-136 (1996).

35. G.J. Tearney, B.E. Bouma, S.A. Boppart, B. Golubovic, E.A. Swanson, and J.G. Fujimoto, "Rapid acquisition of in vivo biological images using optical coherence tomography," Opt. Lett. **21**, 1408-1410 (1996).

36. B. Golubovic, B. E. Bouma, I. P. Bilinsky, J. G. Fujimoto, V. P. Mikhailov, "Ultrathin crystal, room temperature, Cr^{4+}:forsterite laser using near infrared pumping," Opt. Lett. **21**, 1993-1995 (1996).

37. S. Fleischer, B. Pevzner, D.J. Dougherty, E.P. Ippen, M.S. Dresselhaus, and A.F. Hebard, "Phototransformation in visible and near-IR femtosecond pump-probe studies of C_{60} films," Appl. Phys. Lett. **69**, 296-298 (1996).

38. L.E. Nelson, S.B. Fleischer, G. Lenz, and E.P. Ippen, "Efficient frequency doubling of a femtosecond fiber laser," Opt. Lett. **21**, 1759-1761 (1996).

39. S. Namiki, E.P. Ippen, H.A. Haus, and K. Tamura, "Relaxation oscillation behavior in polarization additive pulse mode-locked fiber ring lasers," Appl. Phys. Lett. **26**, 3969-3971 (1996).

40. D.J. Jones, H.A. Haus, and E.P. Ippen, "Subpicosecond solitons in an actively mode-locked fiber laser," Opt, Lett. **21**, 1818-1820 (1996).

41. D.J. Dougherty, S.B. Fleischer, E.L. Warlick, J.L. House, G.S. Petrich, E. Ho, L.A. Kolodziejski, and E.P. Ippen, "Investigation of electric fields, interface charges, and conduction band offsets at ZnSe/GaAs heterojunctions with a novel photoreflectance technique," Proceedings of the Materials Research Society, November 1996.

42. S. Namiki, E.P. Ippen, H.A. Haus, and C.X. Yu, "Energy rate equations for mode-locked lasers," J. Opt. Soc. Am. B.**14**, 1-13 (1997).

43. D.J. Dougherty, S.B. Fleischer, E.L. Warlick, J.L. House, G.S. Petrich, L.A. Kolodziejski, and E.P. Ippen, "Ultrafast carrier dynamics and intervalley scattering in ZnSe," Appl. Phys. Lett., in press (1997).

44. S.A. Boppart, G.J. Tearney, B.E. Bouma, J.F. Southern, M.E. Brezinski, and J.G. Fujimoto, "Noninvasive assessment of the developing xenopus cardiovascular system using optical coherence tomography," Proceedings of the National Academy of Sciences, **94**, 4256-4261 (1997).

45. G.J. Tearney, M.E. Brezinski, B.E. Bouma, S.A. Boppart, C. Pitris, J.F. Southern, and J.G. Fujimoto, "In vivo endoscopic optical biopsy with optical coherence tomography," Science, **276**, 2037-2039 (1997).